VU Research Portal

3D direct impacts of urgan aerosols on dynamics during the CAPITOUL field experiment

Aouizerats, B.; Tulet, P.; Gomes, L.

published in
Geophysical Research Letters
2012

DOI (link to publisher)
10.1029/2012GL053781

document version
Publisher's PDF, also known as Version of record

Link to publication in VU Research Portal

citation for published version (APA)
Aouizerats, B., Tulet, P., & Gomes, L. (2012). 3D direct impacts of urgan aerosols on dynamics during the CAPITOUL field experiment. *Geophysical Research Letters*, *39*, [L23807]. https://doi.org/10.1029/2012GL053781

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal ?

Take down policy
If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

E-mail address:
vuresearchportal.ub@vu.nl

Download date: 04. May. 2023

3D direct impacts of urban aerosols on dynamics during the CAPITOUL field experiment

B. Aouizerats,[1,2] P. Tulet,[1,3] and L. Gomes[1]

Received 6 September 2012; revised 30 October 2012; accepted 5 November 2012; published 14 December 2012.

[1] Evaluating the radiative impacts of aerosol particles is of great interest for understanding atmospheric physics and processes feedbacks. To respond to such objectives, the online fully coupled model Meso-NH is applied to a real case during a two-day Intensive Observation Period (IOP) of the CAPITOUL campaign. The aerosol optical properties are computed from the chemical composition and the size distribution of the particle population, and are compared to observations and analysed at local and regional scales. The differences between two simulations are then studied in order to isolate the direct radiative impacts of aerosols on dynamics. Results show that the aerosol particles generate a forcing on shortwave flux by a decrease of the amount reaching the surface up to 30 Wm^{-2}. The resulting feedbacks lead to a cooling up to 0.6 K on the 2-meter temperature over the city of Toulouse and over the larger 125 km by 125 km area around Toulouse. This cooling is also modeled along the whole boundary layer, leading to a decrease of the boundary layer height up to -50 m during the afternoon and a decrease of the vertical velocities with an average of -3 %. **Citation:** Aouizerats, B., P. Tulet, and L. Gomes (2012), 3D direct impacts of urban aerosols on dynamics during the CAPITOUL field experiment, *Geophys. Res. Lett.*, *39*, L23807, doi:10.1029/2012GL053781.

1. Introduction

[2] Aerosol particles plays a major role in the balance of the Earth-atmosphere space system, and remain a major source of uncertainty in the prediction of climate change due to the large number of external and internal processes modifying the particle properties and their impacts [*Intergovernmental Panel on Climate Change* (*IPCC*), 2007]. Among those impacts, their scattering and absorbing behaviour of solar radiation and the related feedbacks stand as one of the main uncertainties [*Ramanathan et al.*, 1997]. Measurements of the radiative forcings from aerosol particles take place since 40 years and have shown a decrease of 17.6% of the amount of solar radiation reaching the surface for the city of Mexico [*Raga et al.*, 2001]. Other measurements have shown for the Po Valley a decrease of shortwave radiation at ground level between 30 and 70 Wm^{-2} [*Clerici and Melin*, 2008]. This radiative forcings from aerosol particles lead to a cooling of the atmosphere which partly compensate the warming caused by greenhouse gases [*IPCC*, 2007; *Ramanathan and Carmicheal*, 2008]. Recently, aerosol feedbacks have been studied at regional scale during extreme events of dust [*Grini et al.*, 2006] or biomass burning episodes [*Hodzic et al.*, 2007], as well as at climate scales [*Solmon et al.*, 2008]. In this context, this study presents a modeling experiment based on a real-case study aiming at determining the aerosol direct radiative impacts on dynamics in a fine-scale three dimensional configuration during a two-day period over a moderately polluted city.

2. Methodology

2.1. Description of the Situation

[3] This study aims at representing the direct radiative effect of urban aerosols during a two-day real case study. The framework of this paper is the CAPITOUL campaign [*Masson et al.*, 2008]. This field experiment took place from February 2004 to February 2005 in the city of Toulouse, located in southwestern France. This paper focuses on a two-day IOP occurring the 3 and 4 July 2004 during a clear-sky situation. During this period, meteorological as well as aerosol observations were acquired and compared with the model results.

[4] At regional scale, model results show moderately high secondary particle mass concentrations along the plume during day time with mass concentrations at surface level up to 8 $\mu g\ m^{-3}$, while primary aerosols are highly concentrated over the location of cities with mass concentration values up to 6 $\mu g\ m^{-3}$ for Toulouse. The model results (meteorology and aerosols) show similar values as observations acquired during the campaign. A detailed description of the situation as well as comparisons with observations are described by *Aouizerats et al.* [2011].

2.2. Meso-NH: A Fully Coupled Model

[5] To study the direct radiative impacts of aerosol particles, the model Meso-NH [*Lafore et al.*, 1998] is used. Meso-NH is a non-hydrostatic and anelastic atmospheric model jointly developed by CNRM-GAME (Meteo-France/Centre National de Recherche Scientifique) and Laboratoire d'Aérologie (Université Paul Sabatier/Centre National de Recherche Scientifique). In addition to the meteorological variables, Meso-NH computes the gaseous chemistry evolution and solves the aerosol equilibrium at each time step and on each grid point [*Tulet et al.*, 2003] for each of the 3 domains. The chemical reaction module employs a 82 species: ReLACS2 (Reduced Lumped Atmospheric Chemical Scheme 2) scheme [*Tulet et al.*, 2005] based on the CACM (Caltech Atmospheric Chemistry Mechanism) scheme developed by *Griffin et al.* [2002]. The aerosol particle

[1]CNRM-GAME, Météo-France/CNRS, Toulouse, France.
[2]Faculty of Earth and Life Sciences, VU University Amsterdam, Amsterdam, Netherlands.
[3]LACy, Université de La Réunion, Saint-Denis, France.

Corresponding author: B. Aouizerats, Faculty of Earth and Life Sciences, VU University Amsterdam, De Boelelaan 1105, Amsterdam NL-1081 HV, Netherlands. (benjamin.aouizerats@vu.nl)

©2012. American Geophysical Union. All Rights Reserved.
0094-8276/12/2012GL053781

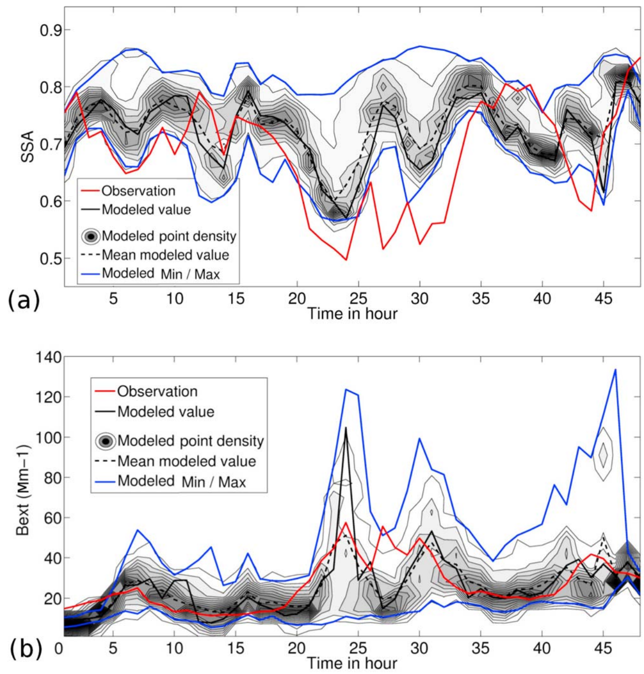

Figure 1. Comparison of (a) single scattering albedo and (b) extinction coefficient observed (in red) and modeled. The comparison is performed over an area of 8 km by 8 km representative of the city of Toulouse. The modeled value of the grid point located on the observation site is drawn in plain black lines. The density function is drawn in grey contours with a 5% step between each contour. The average modeled value for the entire area is drawn in dashed black line. The minimum and maximum values are drawn in plain blue lines.

module, ORILAM-SOA (Organic Inorganic Lognormal Aerosol Model including Secondary Organic Aerosol) [*Tulet et al.*, 2005, 2006] drives the aerosol dynamical processes (coagulation, sedimentation and deposition) as well as the thermodynamical equilibrium between gases and particles along the MPMPO (Model to Predict the Multiphase Partitioning of Organics) scheme [*Griffin et al.*, 2002] for organic species and the scheme EQSAM (EQuilibrium Simplified Aerosol Module) [*Metzger et al.*, 2002] for inorganic species in order to consider the condensation and nucleation processes. The aerosol species considered are: black carbon (BC) and primary organic carbon (OC_p) for the primary species, NO_3^-, SO_4^{2-}, NH_4^+ for the inorganic ions, 10 classes of secondary organic aerosol ($SOA_{1,...,10}$) and water (H_2O). The configuration used for the simulations is made of three nested domains with horizontal resolutions of 10 km, 2.5 km and 500 m, configured with a two-way interaction grid-nesting. More details about the configuration of the simulation can be found in *Aouizerats et al.* [2011]. To consider the radiative impacts of particles, the aerosol optical properties are computed online by the module described by *Aouizerats et al.* [2010].

2.3. Method Used to Analyse the Impacts of Aerosols

[6] In order to isolate the radiative impacts of aerosol particles within this fully coupled approach, the following method is applied. After a two-day spin-up to initialise the atmosphere by the model (including meteorology, chemistry and aerosols), two different simulations are launched from the same initial state. The first one, designated as simulation A, runs without taking into account the radiative impacts of aerosol particles. From the same initial state, a second simulation designated as simulation B is launched, but including the online computation of the aerosol optical properties and then considered as input into the radiation scheme. This module considers the aerosol chemical and physical evolving properties to compute the internally mixed aerosol optical properties at six wavelengths in the shortwave flux from 217.5 nm to 3.19 μm. The feedbacks on dynamical fields are then evaluated by studying the differences.

3. Aerosol Optical Properties: Restitution and Analysis

3.1. Evaluation of the Modeled Aerosol Optical Properties

[7] During the CAPITOUL campaign, an aethalometer measured the absorption coefficient at the wavelength of 550 nm on the downtown site in Toulouse and a nephelometer measured the scattering coefficient at the wavelength of 550 nm on the same location. The nephelometer values are the corrected as described by *Gomes et al.* [2008]. These measured variables can be converted into single scattering albedo and extinction coefficient by the following relations: $B_{ext} = B_{scat} + B_{abs}$ and $SSA = \frac{B_{scat}}{B_{ext}}$ with B_{ext}, B_{scat}, B_{abs} and *SSA* standing for the extinction, scattering absorption coefficients and the single scattering albedo, respectively. Figures 1a and 1b present the comparison of the modeled and observed single scattering albedo and extinction coefficient respectively. The comparison is shown for the observations drawn in plain red line and for the model results corresponding to the 8 by 8 km area standing for the city of Toulouse. The plain black line stands for the modeled values at the grid point corresponding to the observation point location. The grey contours stand for the density of grid points with a 5% step between each contour. the dashed black line stands for the mean value modeled for the Toulouse area. The blue lines stand for the minimum and maximum modeled values in the area. This comparison shows that the model manages to reproduce the evolution of the parameters globally during the two-day IOP.

3.2. Analysis of the Aerosol Optical Properties

[8] Figures 1a and 1b also show the high temporal and spatial variability in the model results. For instance, Figure 1a shows that *SSA* modeled values can reach a minimum of 0.6 and a maximum of 0.85 at the same time (4 UTC on 4 July) on grid points located in the same 8 by 8 km area. In order to link the aerosol optical properties and their spatial variability to the particle composition, Figure 2 presents the horizontal cross sections of *SSA* values over the 2nd and the 3rd domain on 4 July at 4 UTC, where the spatial variability is the highest as shown in Figure 1a. First, we can see that the lowest values of *SSA* over the second and the third domain are located on the location of the city of Toulouse with values reaching 0.6. Over the second domains, the highest values are located on the northwest of Toulouse in the direction of the plume

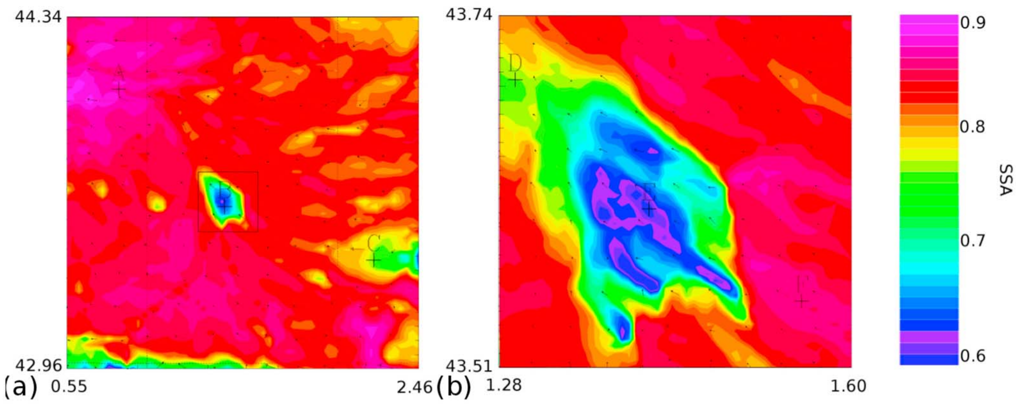

Figure 2. Horizontal cross section of single scattering albedo at 2 meters above ground level over (a) the second domain and (b) the third domain on 4 July 4 UTC.

previously emitted on 3 July and reach values of 0.9. Table 1 presents the chemical composition of aerosols located on six points and marked by black crosses in Figure 2. This table helps to link the *SSA* values with the chemical composition. Indeed, the lowest values of *SSA* as for point B and E stand for high ratios of primary aerosols (71% and 81% respectively), while highest values of *SSA* as for point A and F stand for high ratios of secondary aerosols (78% and 81% respectively). The location of primary particles matches with the location of high anthropical emissions while secondary particles are usually formed along the advected plume. In this case, the city of Toulouse concentrates a large amount of emissions and therefore the location of the lowest *SSA* values.

4. Radiative Impacts at the Surface

[9] The differences between simulation A and simulation B are investigated. The aerosol optical depth is increasing over the 48 hours of simulation, with moderate values and a maximum of 0.13 at the end of the simulation. The difference of shortwave flux reaching the surface between the two simulations at the location of the downtown site ($SW_B - SW_A$) is also investigated. The maximum of differences occurs at 13 UTC on 4 July with a value of 30 Wm^{-2} corresponding of 5% of the total shortwave flux reaching the surface at this moment. Those results are illustrated in the auxiliary material.[1]

4.1. Local Impacts Over Toulouse

[10] The difference of radiative forcings leads to feedbacks on dynamics. First, the impacts on the two-meter temperature are studied over the third domain. Figure 3a presents the evolution of the difference of the two-meter temperature between the two simulations over the third domain ($T_{2m,B} - T_{2m,A}$). The density function is drawn in grey contours with a 5% step between each contour and the average value is drawn in plain red line. Figure 3a shows that the averaged two-meter temperature is cooler on simulation B than in simulation A over the third domain. The density function shows that there is a large dispersion of the values especially during the night, leading to a noisy signal. However, the average value shows that there are only 4 points on 48 which present positive values, and 2 of them occur at the very end of the simulation. The average value shows a cooling of the two-meter temperature due to aerosols up to 0.3 K. The positive values at the end of the simulation do not stand for aerosol radiative impact but from a drift between the two simulations leading to different locations of dynamical structures. To get rid of this constraint, the radiative impacts of aerosol particles are then investigated over the second domain which includes a larger area and the outgoing plume from Toulouse.

4.2. Regional Impacts Over the Second Domain

[11] Figure 3b presents the evolution of the difference of the two-meter temperature between the two simulations over the second domain ($T_{2m,B} - T_{2m,A}$). As in Figure 3a, the density function is drawn in grey contours with a 5% step between each contour and the average value is drawn in plain red line. It should be noted that most of secondary aerosols from the Toulouse plume are fromed away from emissions and are present over the second domain. The cooling occurring over the third domain is still more pronounced over the whole Toulouse region. The signal presents almost no noise, and the dispersion is closer to the averaged value. Almost all points show a cooling of the two-meter temperature due to aerosol particles, and the average value presents a maximum of cooling at the end of the simulation of 0.6 K. Finally, Figure 3c reproduces the evolution of the two-meter temperature difference averaged over the third domain in red and over the second domain in blue. Except for the last 6 hours, the pattern of the evolution of the two-meter temperature difference shows the same pattern. The radiative impacts of aerosol particles seem to show

Table 1. Single Scattering Albedo Values and Aerosol Mass Composition Associated With the Six Points Reported in Figure 2

Point	*SSA*	OC_p (%)	*BC* (%)	NO_3^- (%)	SO_4^{2-} (%)	NH_4^+ (%)	*SOA* (%)	H_2O (%)
A	0.86	18	4	12	9	7	8	42
B	0.64	59	12	9	4	4	5	7
C	0.75	22	5	16	12	9	17	19
D	0.76	30	7	19	8	9	9	18
E	0.59	67	14	5	3	3	4	4
F	0.86	15	4	19	9	9	8	36

[1]Auxiliary materials are available in the HTML. doi:10.1029/2012GL053781.

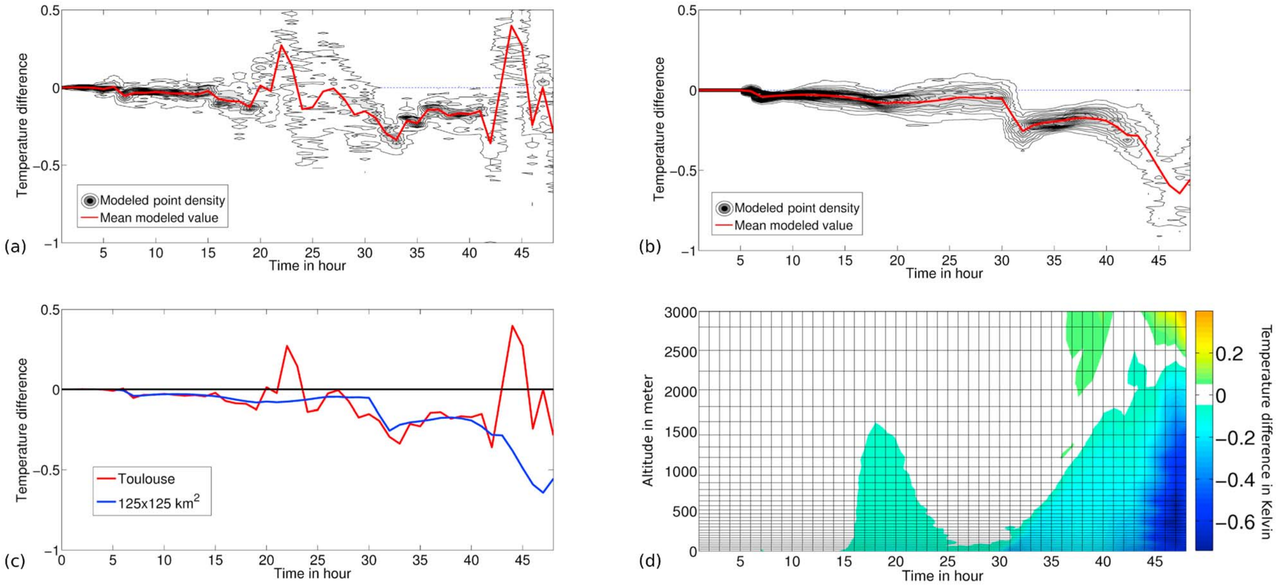

Figure 3. Evolution of the difference of 2-meter temperature between the two simulations over (a) the third domain and (b) the second domain. The density function is drawn in grey contours with a 5% step between each contour and the average value is drawn in plain red line. (c) Average values. (d) The evolution of the 2-meter differences profiles averaged over the second domain.

similar results over the second domain and over the third domain. However the investigation of results over the second domain allows the signal not to be disrupted by local impacts on the location of dynamical structures and seems to give more readable interpretation.

5. Radiative Impacts in the Boundary Layer

[12] To study the radiative impacts of aerosol particles on the boundary layer, results are investigated over the second domain. Figure 3d shows the evolution of the difference of temperatures profiles between the two simulations ($T_{pr,B} - T_{pr,A}$). This difference profile is the average of the values over the second domain. Figure 3d shows that the cooling occurring at the altitude of two-meter above ground level also occurs along the whole boundary layer. Indeed, a net cooling due to aerosol particles occurs on 3 July from 15 UTC with values between 0.05 and 0.15 K. The vertical structure of this cooling follows the vertical development of the boundary layer. On 4 July, the same pattern occurs with a cooling along the whole boundary layer, but with higher values up to 0.6 K at the end of the simulation. The aerosol impacts on dynamics is then a cooling along the whole boundary layer, due to the difference of shortwave flux reaching the surface and scattered and absorbed by particles. The difference of shortwave flux seem predominant to the feedbacks on dynamics on the lower atmosphere, even with low single scattering albedo values as shown in Figure 1a.

[13] The radiative impacts of aerosol particles also occur on other meteorological parameters. First, the negative difference of energy reaching the boundary layer leads to a decrease of the entrainment. The absolute values of vertical wind velocities are lower by 3% during the two days of simulation in the boundary layer, with a maximum of 5% during the afternoon on 4 July. This lower entrainment leads then to a lower expansion of the boundary layer up to 50 meters on July 4 at 18 UTC, corresponding to 2.5% of the total boundary layer height.

6. Conclusions

[14] The fully coupled model Meso-NH has been used to isolate and estimate the direct radiative effects of aerosol particles for a two-day period occurring during the CAPITOUL field experiment. The specific situation during this IOP showed a clear-sky coverage as well as a recirculation of Toulouse aerosol plume within the second domain. First, the online modeled aerosol optical properties have been compared to observations and show consistent values as well as a high temporal and spatial variability. The modeled aerosol optical properties were also analysed and linked with the modeled aerosol chemical composition. The direct radiative forcings from aerosols have been evaluated and show a decrease of the total amount of solar flux reaching the surface up to 30 Wm^{-2}. The resulting feedbacks on dynamics have been investigated and show a net cooling of the two-meter temperature over the whole Toulouse region corresponding to a 125 km by 125 area km. This cooling is maximum at the end of the simulation with a maximum of 0.6 K. Then, it has been shown that the cooling occurs along the whole boundary layer, leading to a decrease of the intensity of the vertical wind velocities up to 5% and a decrease of the top of the boundary layer height up top 50 m. Consequently this study highlights the role of the aerosol particles on meteorology fields, even in moderately low polluted conditions as in the studied case. It is legitimate to wonder if the values of the modeled cooling would be the same in largely polluted cities. One of the main question remaining stands for the trend of the cooling during the simulation which tends to continuously increase. The high computer time required for such simulations did not allow us to continue the simulations in

order to investigate how impacts on temperature behave for a longer period, or within an ensemble of runs.

[15] **Acknowledgments.** The authors are indebted to Valery Masson for the organization of the CAPITOUL experiment. They also thank Marc Mallet for his advice. They gratefully thank Meteo France for its support. Finally, all my thoughts go to Laurent.

[16] The Editor thanks two anonymous reviewers for assistance evaluating this paper.

References

Aouizerats, B., O. Thouron, P. Tulet, M. Mallet, L. Gomes, and J. S. Henzing (2010), Development of an online radiative module for the computation of aerosol optical properties in 3-D atmospheric models: Validation during the EUCAARI campaign, *Geosci. Model Dev.*, *3*, 553–564, doi:10.5194/gmd-3-553-2010.

Aouizerats, B., P. Tulet, G. Pigeon, V. Masson, and L. Gomes (2011), High resolution modelling of aerosol dispersion regimes during the CAPITOUL field experiment: From regional to local scale interactions, *Atmos. Chem. Phys.*, *11*, 7547–7560, doi:10.5194/acp-11-7547-2011.

Clerici, M., and F. Melin (2008), Aerosol direct radiative effect in the Po Valley region derived from AERONET measurements, *Atmos. Chem. Phys.*, *8*, 4925–4946.

Griffin, R. J., D. Dabdub, and J. H. Seinfeld (2002), Secondary organic aerosol: 1. Atmospheric chemical mechanism for production of molecular constituents, *J. Geophys. Res.*, *107*(D17), 4332, doi:10.1029/2001JD000541.

Grini, A., P. Tulet, and L. Gomes (2006), Dusty weather forecasts using the MesoNH mesoscale atmospheric model, *J. Geophys. Res.*, *111*, D19205, doi:10.1029/2005JD007007.

Gomes, L., M. Mallet, J. C. Roger, and P. Dubuisson (2008), Effects of the physical and optical properties of urban aerosols measured during the CAPITOUL summer campaign on the local direct radiative forcing, *Meteorol. Atmos. Phys.*, *108*, 289–306.

Hodzic, A., S. Madronich, B. Bohn, S. Massie, L. Menut, and C. Wiedinmyer (2007), Wildfire particulate matter in Europe during summer 2003: Meso-scale modeling of smoke emissions, transport and radiative effects, *Atmos. Chem. Phys.*, *7*, 4043–4064.

Intergovernmental Panel on Climate Change (IPCC) (2007), *Climate Change 2007: The Scientific Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., Cambridge Univ. Press, Cambridge, U. K.

Lafore, J., et al. (1998), The Meso-NH atmospheric simulation system. Part I: adiabatic formulation and control simulations, *Ann. Geophys.*, *16*, 90–109.

Masson, V., et al. (2008), The Canopy and Aerosol Particles Interactions in TOulouse Urban Layer (CAPITOUL) experiment, *Meteorol. Atmos. Phys.*, *102*, 135–157.

Metzger, S., F. Dentener, S. Pandis, and J. Lelieveld (2002), Gas/aerosol partitioning: 1. A computationally efficient model, *J. Geophys. Res.*, *107*(D16), 4312, doi:10.1029/2001JD001102.

Raga, G. B., D. B. T. Castro, A. Martinez-Arroyo, and R. Navarro-Gonzalez (2001), Mexico City air quality: A qualitative review of gas and aerosol measurements (1960–2000), *Atmos. Environ.*, *35*, 4041–4058.

Ramanathan, V., and G. Carmicheal (2008), Global and regional climate changes due to black carbon, *Nat. Geosci.*, *1*, 4221–4227.

Ramanathan, V. R., et al. (1997), A multi-agency proposal for a field experiment in the Indian Ocean, *C4 Publ. 162*, Cent. for Clouds, Chem. and Clim., Scripps Inst. of Oceanogr., Univ. of Calif., San Diego.

Solmon, F., M. Mallet, N. Elguindi, F. Giorgi, A. Zakey, and A. Konar (2008), Dust aerosol impact on regional precipitation over western Africa, mechanisms and sensitivity to absorption properties, *Geophys. Res. Lett.*, *35*, L24705, doi:10.1029/2008GL035900.

Tulet, P., V. Crassier, F. Solmon, D. Guedalia, and R. Rosset (2003), Description of the Mesoscale Nonhydrostatic Chemistry model and application to a transboundary pollution episode between northern France and southern England, *J. Geophys. Res.*, *108*(D1), 4021, doi:10.1029/2000JD000301.

Tulet, P., V. Crassier, F. Cousin, K. Suhre, and R. Rosset (2005), ORILAM, a three-moment lognormal aerosol scheme for mesoscale atmospheric model: Online coupling into the Meso-NH-C model and validation on the Escompte campaign, *J. Geophys. Res.*, *110*, D18201, doi:10.1029/2004JD005716.

Tulet, P., A. Grini, R. J. Griffin, and S. Petitcol (2006), ORILAM-SOA: A computationally efficient model for predicting secondary organic aerosols in three-dimensional atmospheric models, *J. Geophys. Res.*, *111*, D23208, doi:10.1029/2006JD007152.